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Atmospheric deposition of nitrogen and sulfur over southern Europe with focus on the Mediterranean and the Black Sea

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HIGHLIGHTS

- Atmospheric N and S depositions over Mediterranean and Black seas are simulated.
- N transported from upwind sources is deposited over the Mediterranean.
- Dry deposition dominates over wet deposition in Mediterranean and Black Sea.
- Atmospheric N inputs are comparable to N export in Black and W. Mediterranean seas.
- Atmospheric N input exceeds the N export in the East Mediterranean Sea.

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ABSTRACT

Atmospheric deposition provides significant amounts of nutrients to the continental and marine ecosystems. Using the mesoscale WRF/CMAQ modeling system, the nitrogen (N) and sulfur (S) atmospheric deposition fluxes over the Mediterranean and the Black seas and continental Europe are evaluated for the year 2008. The annual N and S deposition fluxes are calculated to be 4.89 Tg(N) yr^{-1} and 2.07 Tg(S) yr^{-1} over continental Europe, 0.92 Tg(N) yr^{-1} and 0.52 Tg(S) yr^{-1} over West Mediterranean, 1.10 Tg(N) yr^{-1} and 0.84 Tg(S) yr^{-1} over East Mediterranean and 0.36 Tg(N) yr^{-1} and 0.17 Tg(S) yr^{-1} over the Black Sea. Inorganic N deposition fluxes are calculated to be about 3 times higher than gaseous organic N deposition fluxes. Comparison to available observations associates the annual mean model estimates with about 40 ± 30% of uncertainty depending on location. Dry deposition dominates over wet deposition for both N and S in agreement with the observations. Results suggest that an important fraction of the N deposited over the Mediterranean basin can be attributed to transported N species while S deposition is dependent more on the local emissions. In Black Sea and West Mediterranean Sea waters the calculated atmospheric N inputs are comparable to the N export measured by sediment traps whereas in the East Mediterranean N input exceeds by a factor of about 5 the N export. Our simulations show that the critical N load of 1 g(N) m^{-2} yr^{-1} is exceeded over 84% of the European forested areas.

1. Introduction

Atmospheric deposition of nitrogen (N) and sulfur (S) species from both natural and anthropogenic sources lead either to benefits (fertilization) or drawbacks (acidification and accumulation of excess nutrients) for the ecosystems (Driscoll et al., 2003). With the advent of the Anthropocene the transfer of these species has increased over natural levels, modifying thus their biogeochemical cycles in both terrestrial and aquatic ecosystems (Galloway et al., 2008). First estimates indicate that the human-induced increase in atmospheric N deposition to the oceans may account globally for up to ~3% of the annual new oceanic primary productivity (Duce et al., 2008). Especially for semi-enclosed marine ecosystems such as the Mediterranean Sea, atmospheric deposition of N may account for up to 35–60% of new production (Christodoulaki et al., 2013). Furthermore, in the atmosphere, ozone (O_3) production is driven by nitrogen oxide (NO_x) availability and atmospheric acidity

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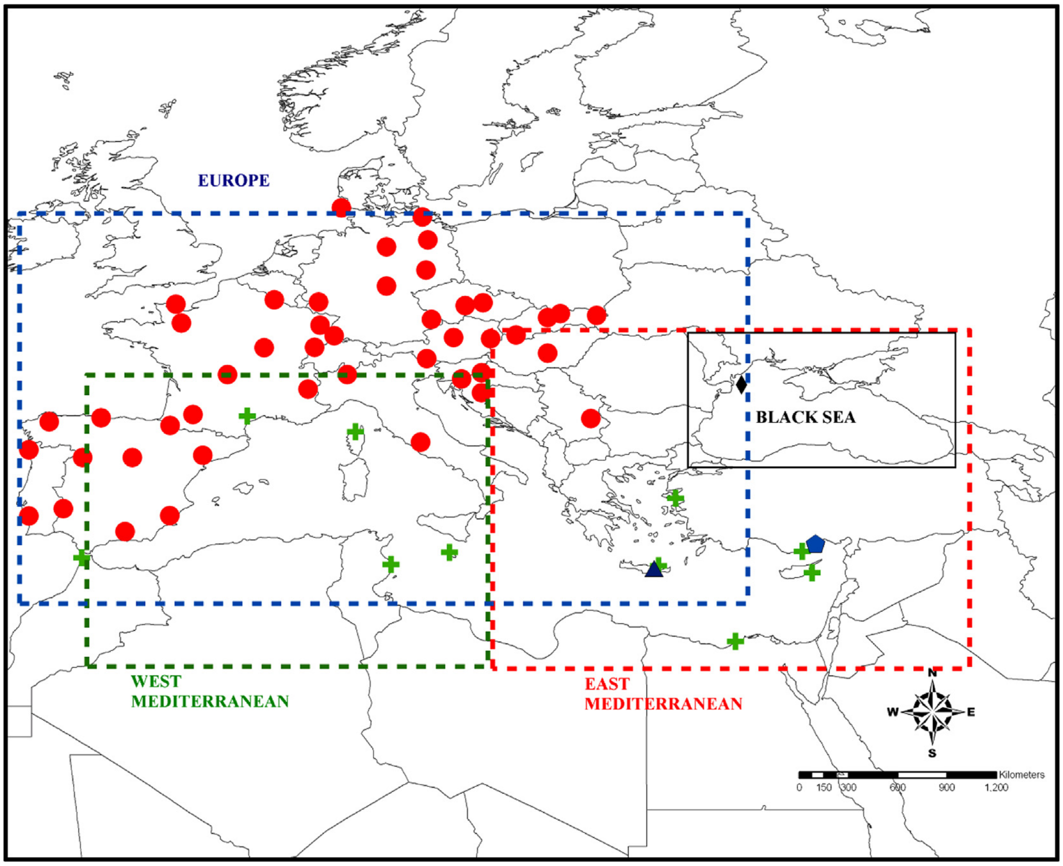

Fig. 1. The locations of the EMEP wet deposition stations (circles) and Finokalia station for dry deposition (triangle). Crosses represent monitoring stations in Markaki et al. (2010), diamond represents the monitoring station at Zmiinyi Island, Ukraine (Medinets and Medinets, 2012) and pentagon represents the monitoring station in Erdemli, Turkey (Kocak et al., 2010). The model domain extents from 18.98°N, 3.58°W to 49.82°N, 57.64°E. The EUROPE box represents Europe (from 29.95°N, 7.47°W to 54.98°N, 32.03°E), the East Mediterranean box represents East Mediterranean (from 41.31°N, 27.67°E to 46.37°N, 43.76°E) and the West Mediterranean box represents Western Mediterranean (from 27.60°N, 2.77°W to 45.17°N, 15.14°E). The Black Sea box extends from 41.31°N, 27.67°E to 46.37°N, 43.76°E.

is controlled by nitric acid (HNO_3) and sulfuric acid (H_2SO_4) formation (Seinfeld and Pandis, 2006). Thus, deposition of atmospheric N and S species impacts on atmospheric chemistry. There are different projections for the future levels of N and S deposition fluxes over Europe based on different emission scenarios in the literature that indicate control of the emissions of SO_2 and NO_x but not those of NH_3 (Dentener et al., 2006; Geels et al., 2012; Lamarque et al., 2013).

Monitoring of deposition, particularly dry deposition, is very challenging, especially over water bodies (Pryor et al., 2008), and can provide data of only limited geographical coverage (Fowler et al., 2009). Because deposition depends on surface characteristics (dry deposition) and precipitation rates (wet removal), significant interpolation errors can occur when deposition is to be evaluated over large areas based on observations. Atmospheric chemistry and transport models (CTMs) are unique tools to provide integrated view of the temporal and spatial variations of dry and wet deposition over local to global scales. CTMs can calculate critical loads in order to provide estimates of the environmental impact of deposition. They can use future emission estimates to assess environmental change and provide advice to policy makers. Such model output requires extensive evaluation by comparison to observed deposition fluxes (Flechard et al., 2011).

Simpson et al. (2006) European Monitoring and Evaluation Programme (EMEP) mesoscale modeling study calculated that dry deposition of reduced N species is the major contributor to total reactive N deposition in Central and southern Europe. Menegoz et al. (2009) global CTM simulations of sulfate ($SO_4^=$) compared with EMEP observations over Europe showed overestimations in surface atmospheric $SO_4^=$ levels that have been attributed to underestimated wet deposition.

Despite the importance of atmospheric deposition for ecosystems, there is a limited number of modeling studies dedicated to the evaluation of atmospheric deposition of N and S to the Mediterranean and the Black Seas (e.g. Markaki et al., 2010; references therein, Medinets and Medinets, 2012). The present mesoscale modeling study aims to fill this gap by performing, the first to our knowledge study of one full year mesoscale simulation of atmospheric deposition of N and S over Europe, with special focus on the Mediterranean and Black Sea.

2. Materials and methods

2.1. The modeling system

The Advanced Research Weather Research and Forecasting mesoscale meteorological model (WRF-ARW v3.1.1; Skamarock and Klemp, 2008) has been used to calculate the meteorological fields necessary to drive the Community Multiscale Air Quality (CMAQ) model, v4.7 (Byun and Schere, 2006). The model domain (Fig. 1) covers most of the Europe, North Africa and the Middle East (from 18.98°N, 3.58°W to 49.82°N, 57.64°E) on a 30 × 30 km horizontal resolution, extending up to ~16 km on 23 vertical levels. The initial and boundary meteorological conditions for the WRF model have